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Correlated electron transport through a quantum dot: The multiple-level effect

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Electron transport through a multilevel quantum dot with intradot Coulomb repulsion is discussed theoretically. Based on the calculated local density of states, we argue that characteristic peaks should be seen in differential conductance if the dot has a group of closely spaced levels. These peaks provide a hallmark of Kondo-type correlation. Our proposal is especially pertinent if one realizes that Kondo temperature in a multilevel system may be enhanced by orders of magnitude over that for a single-level system.

The role of intradot Coulomb repulsion in resonant tunneling through a quantum dot has been the subject of strong interest recently.[1–5] On the assumption that the dot has only one (spin-degenerate) level, linear-response conductance at $T = 0$ was studied as a function of the level energy E_0, and was shown to be nearly constant[1,3] ($= 2e^2/h$) in the range $E_F - U \leq E_0 \leq E_F$ due to Kondo resonance that develops near the Fermi level E_F. (Here U is the intradot Coulomb repulsion energy.) This is in marked contrast to the case of high temperatures (roughly speaking, $T \gtrsim 5T_K$ where T_K is the so-called Kondo temperature), where the Kondo resonance is lost and two separate peaks are seen at $E_0 = E_F - U$ and E_F. However, it was also argued[2] that at low but experimentally accessible temperatures, this remarkable plateau-like structure is lost and one would observe, instead, two separate peaks similar to those at high temperatures, the effect of Kondo-type correlation being merely to add shoulders. This is because T_K varies with E_0: it decreases as E_0 increases (decreases) from $E_F - U$ (E_F), i.e., as one goes from the valence fluctuation regime (the average number of electrons in the dot $N_{\rm dot}$ deviates from 1) to the Kondo regime ($N_{\rm dot} \approx 1$).

In the Anderson model with $U = \infty$ for a single-level dot, $k_B T_K = \sqrt{W\Delta}\exp(\pi E_0/2\Delta)$ in the Kondo regime,[6,7] where Δ is the level width due to tunneling, and E_0 is measured relative to E_F. Furthermore, W is a cutoff of the order of $\min(B, U)$, where B is half the bandwidth of the leads. In case both the dot and the leads are made of GaAs (n-modulation doping in the leads), typical parameters are $W = 5$ meV, $\Delta = 0.1$ meV, and $E_0 = -0.5$ meV. Using these values in the above formula, one obtains $T_K = 3$ μK, which is low enough to discourage the experimentalist. Fortunately, this is not the whole story. In the present paper, we will argue that multiple-level effects, which have been overlooked so far, are crucially important in real quantum dots: their inclusion may enhance T_K by orders of magnitude. More importantly, if the dot has a multiple-level structure, characteristic peaks appear in the differential conductance-bias voltage curve, which should provide a hallmark of the Kondo-type correlation in quantum-dot systems.[8]

As a starting point of our discussion, let us consider, for simplicity, a dot which is coupled predominantly with the left lead, the coupling with the right lead being neglected for the moment. (This is not unrealistic since the Kondo effect depends exponentially on the dot-lead coupling. A slight asymmetry results in predominant coupling with one lead.) The Anderson Hamiltonian describing this system is

$$H = \sum_{k,\sigma} \varepsilon_k c^\dagger_{k\sigma} c_{k\sigma} + \sum_{k,m,\sigma} \left(V_k b^\dagger_{m\sigma} c_{k\sigma} + \text{H.c.}\right) + H_{\rm dot}, \tag{1}$$

$$H_{\rm dot} = \sum_{m,\sigma} \varepsilon_m b^\dagger_{m\sigma} b_{m\sigma} + U \sum_{m,m'} n_{m\uparrow} n_{m'\downarrow}. \tag{2}$$

Here the annihilation operators $c_{k\sigma}$ and $b_{m\sigma}$ correspond to an electron in the (left) lead with wave vector k (energy ε_k) and an electron in the mth dot level (energy ε_m), respectively, with spin σ. It is assumed that the dot levels are orbitally nondegenerate.

Let us now consider an isolated dot described by $H_{\rm dot}$. Suppose it has $2M$ electrons occupying the lowest M levels and the dot levels are designated, in the order of increasing energy, as $m = -M, -M+1, -M+2, \ldots, -1, 0, 1, \ldots$. Namely, the lowest unoccupied level is $m = 0$. If we add an electron to an unoccupied level $m = \alpha$ (≥ 0), the increase in the total energy is $E_\alpha \equiv \varepsilon_\alpha + 2MU$. Further addition of an electron to level β (≥ 0) increases the total energy by $\varepsilon_\beta + (2M+1)U =$

$E_\beta + U$. If the lowest M levels are always occupied, we can replace Eq. (2) with

$$H_{\rm dot} = \sum_{m,\sigma} E_m b^\dagger_{m\sigma} b_{m\sigma} + U \sum_{m,m'} n_{m\uparrow} n_{m'\downarrow}, \qquad (2')$$

where $m, m' = 0, 1, 2, \ldots$. In the situation we are concerned with, those levels $m \geq 0$ satisfying $E_m < 0$ contribute to the correlation effect and so must be retained in the summation in Eq. (2') (Fig. 1).

The importance of multiple-level effects depends on whether U is larger or smaller than the level separation. Figure 2 presents U and the average level separation δ (i.e., the average of $E_m - E_{m-1}$) for both electrons and holes in a GaAs dot as a function of its radius R. Here, the estimation was made using $U \simeq 1/\kappa R$ ($\kappa = 13$ is the dielectric constant) and $\delta = 2\hbar^2/m^* R^2$ ($m^* = 0.067$ and 0.45 for electrons and heavy holes, respectively). It is seen that $U \gg \delta$ except very small dots ($R \lesssim 500$ Å for electrons and $R \lesssim 100$ Å for holes), so real dots should be treated as a multiple-level system in discussing correlation effects. The Kondo temperature T_K for a multiple-level dot can be estimated from[9]

$$T_K = \left(\prod_i \frac{W}{T_K + (E_i - E_0)} \right) T_K^{(1)}, \qquad (3)$$

where $T_K^{(1)}$ is the Kondo temperature with level E_0 alone, $W \gtrsim \min(B, U)$ as before and i ranges over those levels satisfying $E_i < 0$. As far as $T_K \ll W$ and $E_i - E_0 \ll W$, the factor in the large parentheses in Eq. (3) may enhance $T_K^{(1)}$ by several orders of magnitude, making T_K an experimentally accessible value.

When voltage V is applied between the two leads, current I flows through the dot, which at $T = 0$ can be expressed as[10]

$$I = \frac{2e}{\hbar} \frac{\Delta^R \Delta^L}{\Delta^R + \Delta^L} \int_0^{eV} d\varepsilon \, D(\varepsilon), \qquad (4)$$

where $\Delta(\varepsilon) \equiv \pi \sum_k V_k^2 \delta(\varepsilon - \varepsilon_k)$ is assumed to be independent of ε (L and R denoting coupling with the left and right leads, respectively), and the Fermi level of the left lead is set equal to 0. Also $D(\varepsilon)$ is the local density of states for one spin, written as[7,11]

$$D(\varepsilon) = D^<(\varepsilon) + D^>(\varepsilon) \qquad (5)$$

$$= -\frac{1}{\pi} \mathrm{Im} \sum_i \left\langle g \left| b^\dagger_{i\sigma} \frac{1}{\varepsilon^+ - \bar{E} + H} b_{i\sigma} \right| g \right\rangle$$

$$-\frac{1}{\pi} \mathrm{Im} \sum_i \left\langle g \left| b_{i\sigma} \frac{1}{\varepsilon^+ + \bar{E} - H} b^\dagger_{i\sigma} \right| g \right\rangle. \qquad (6)$$

Here $|g\rangle$ and $\bar{E}$ are the ground state wave function and eigenvalue, respectively [$H|g\rangle = \bar{E}|g\rangle$ with H given by Eqs. (1) and (2')], $\varepsilon^+ \equiv \varepsilon + i0^+$, and $D^<(\varepsilon)$ and $D^>(\varepsilon)$ are the density of filled and empty states, respectively. [$D(\varepsilon)$ is independent of σ if we assume that the ground state is a spin singlet.]

We evaluate $D(\varepsilon)$ for a two-level dot with $U = \infty$ by the variational method by Gunnarsson and Schönhammer.[11] In obtaining $|g\rangle$, we diagonalize H in the space $|0\rangle \oplus |1,k\rangle \oplus |2,k\rangle$ ($k < k_F$, the Fermi wave vector) where $|0\rangle$ is the filled Fermi sea (of the left lead) and $|i,k\rangle \equiv (1/\sqrt{2}) \sum_\sigma b^\dagger_{i\sigma} c_{k\sigma} |0\rangle$. Furthermore, $b_{i\sigma}|g\rangle$ is limited to the space spanned by $c_{k\sigma}|0\rangle$ and $(1/\sqrt{2}) \sum_\sigma b^\dagger_{i\sigma} c_{p\sigma} c_{k\sigma} |0\rangle$ ($p, k < k_F$), and $b^\dagger_{i\sigma}|g\rangle$ is limited to the space spanned by $b^\dagger_{i\sigma}|0\rangle$ and $c^\dagger_{k\sigma}|0\rangle$ ($k > k_F$). These approximations lead to

$$D^<(\varepsilon) = \left(-\frac{1}{2\pi} \right) \mathrm{Im} \sum_{i,k} \frac{B_{ik}^2}{\varepsilon^+ - \delta E - \varepsilon_k - 2 \sum_{i,p<k_F} \frac{V_p^2}{\varepsilon^+ - \delta E - \varepsilon_p - \varepsilon_k + E_i}}, \qquad (7)$$

$$D^>(\varepsilon) = \left(-\frac{1}{\pi} \right) A^2 \mathrm{Im} \frac{g_1(\varepsilon) + g_2(\varepsilon)}{g_1(\varepsilon) g_2(\varepsilon) - h(\varepsilon)^2}, \qquad (8)$$

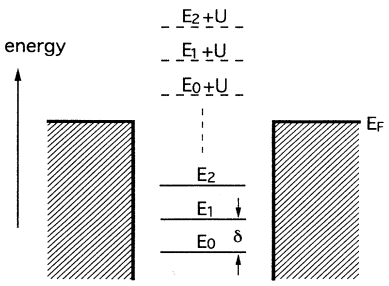

FIG. 1. Schematic energy diagram of a dot coupled to two leads via tunneling. Disregarding the fully occupied levels as explained in the text, the energy levels of the first and second added electrons are indicated by solid and dashed lines, respectively.

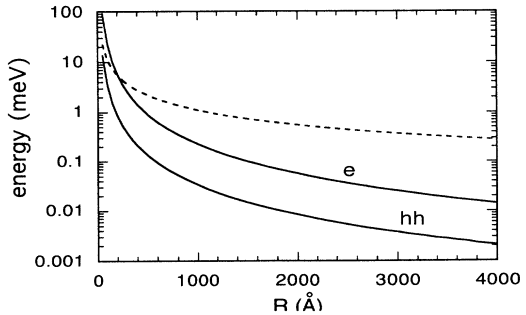

FIG. 2. Coulomb repulsion energy U (dashed line) and average level separation δ (solid lines) as functions of radius R for a disk-shaped GaAs dot. The δ is shown for both electrons (e) and heavy holes (hh).

with $h(\varepsilon) \equiv \sum_{Q>k_F}(V_Q^2/\varepsilon^+ + \delta E - \varepsilon_Q)$ and $g_i(\varepsilon) \equiv \varepsilon^+ + \delta E - E_i - h(\varepsilon)$. Furthermore, A and B_{ik} in Eqs. (7) and (8) are the coefficients of $|0\rangle$ and $|i,k\rangle$, respectively, in $|g\rangle$, and $\delta E \equiv \bar{E} - \langle 0|H|0\rangle$.

Figure 3 presents the calculated $D(\varepsilon)$ for different values of E_0, with the level separation $\delta = E_1 - E_0$ kept constant at 0.2 meV. The parameters used simulate *electron* transport through a GaAs dot with $R \approx 1000$ Å. Aside from the sharp Kondo resonance at the Fermi level $\varepsilon = 0$, two side peaks located roughly at $\pm\delta$ are clearly seen. These side peaks have been obtained by several different methods:[12] The lower side peak, for example, originates from the transfer of a dot electron in the ground level to the lead which makes possible the transfer of a conduction electron in the lead to the upper dot level. Figure 3 clearly shows that the lower side peak, which is orders of magnitude stronger than the upper side peak for small enough E_0 [Kondo regime, Fig. 3(a)], loses intensity and gains width as E_0 goes up [going into the valence fluctuation regime, Fig. 3(b)], until it is negligibly small compared to the upper side peak [Fig. 3(c)].

Since current I is related to $D(\varepsilon)$ by Eq. (4), differential conductance dI/dV at finite V is proportional to $D(eV)$. Thus differential conductance as a function of V should exhibit similar peaks as $D(\varepsilon)$. If E_0 is set in the valence fluctuation regime, where both side peaks are seen in $D(\varepsilon)$, dI/dV should exhibit peaks for both polarities of V. In the Kondo regime, where one of the side peaks is dominant in $D(\varepsilon)$, dI/dV should exhibit a peak for only one polarity of V. These peaks in differential conductance should provide direct evidence for the much sought after Kondo-type correlation.

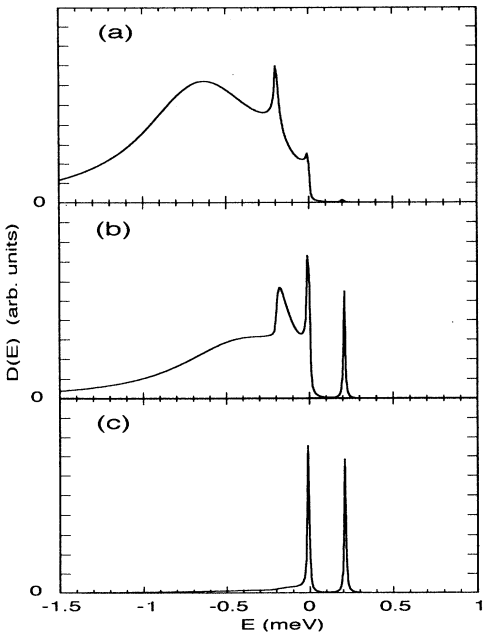

FIG. 3. Calculated density of states $D(\varepsilon)$ for various values of E_0: (a) -0.8 meV, (b) -0.6 meV, and (c) -0.4 meV. The level separation δ is kept constant at 0.2 meV. Other parameters are $\Delta = 0.17$ meV, B (band half-width of the lead)$= 3$ meV, and the imaginary part of ε is 5 μeV.

These side peaks in $D(\varepsilon)$ are known to survive up to higher temperatures than the Kondo peak.[12] This, combined with the multiple-level enhancement of T_K [Eq. (3)], makes our proposed scheme of using differential conductance very promising.

In Fig. 3, the upper side peak is much sharper than the lower side peak. This should not be taken too seriously because our $D^<(\varepsilon)$ (which gives rise to the lower side peak) is correct to order $1/N$ in the $1/N$ expansion but $D^>(\varepsilon)$ is correct only up to the zeroth order of $1/N$ and less accurate,[13] where N is the degeneracy of the dot levels (2 in our case). To treat both side peaks to order $1/N$, a larger basis set is required for $D^>(\varepsilon)$, which is not pursued here.

In order to observe the predicted peaks in dI/dV, it is required that $\delta \ll U$ and at the same time δ should be sufficiently large that the side peaks are clearly distinguishable from the Kondo peak. In the case of electron transport, we estimate that the appropriate dot size is $R \lesssim 1000$ Å (Fig. 2). It should, however, be remembered that δ plotted in Fig. 2 is the *average* level spacing. Even in a small dot where average δ is comparable to U, there always exist groups of levels with much smaller spacing than the average. This is especially true if the dot is highly symmetrical. It is known that the electronic energy spectrum of a dot has an abundance of high angular momentum states.[14,15] A slight deviation of the dot shape from circular would split these degenerate levels. Therefore, in view of the large U (which favors Kondo-type correlation) in small dots, dots with $R \approx 200$ Å (for electrons) are also promising. Another interesting possibility is to study *hole* transport. Since the average δ for holes is an order of magnitude smaller than that of electrons (Fig. 2), one may utilize both the large U and small δ if $R \approx 500$ Å.

Recently, a problem somewhat similar to ours was taken up in Ref. 5 which studied Zeeman splitting-induced change of transport through a dot in the context of the Kondo effect. In the Zeeman splitting case, there is no Kondo resonance in $D(\varepsilon)$: The original Kondo resonance at the Fermi level is *split* by a magnetic field into two peaks. On the other hand, in the case of many spin-degenerate levels, side peaks appear *in addition to* the Kondo resonance. The Zeeman splitting is interesting in its own right; however its observation should still require cooling down the system to a very low temperature, something which may not be easy to achieve. The multiple-level case we have studied is pertinent to nearly all the real situations, since real dots are inherently multiple-level systems.

We have limited our discussion to the case of asymmetric coupling. When the coupling with both leads is important and voltage is applied between them, generally a Kondo resonance and side peaks attached to *each* of the two Fermi levels occur,[5,8] and peaks would be seen in dI/dV as in the asymmetric case. However, our preliminary result[8] indicates that a different situation may arise for this symmetric coupling, a detailed account of which will be reported elsewhere.[16]

Finally, it should also be interesting to study the con-

ductance of a (continuous) quantum wire to which a quantum dot, placed adjacent to but apart from it, is coupled via tunneling. This configuration simulates the conventional Kondo effect in that the dot acts as a scatterer with the Kondo resonance suppressing, rather than enhancing, the conductance.

In summary, we have proposed a scheme of identifying Kondo-type correlation in transport through a multiple-level dot that utilizes differential conductance as a function of the voltage applied between the leads. We hope that our work will encourage more experimental efforts in this fascinating field.

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